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Receptor modelling of both particle composition and size distribution from a background site in London, UK

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Abstract

Positive Matrix Factorisation (PMF) analysis was applied to PM_{10} chemical composition and particle Number Size Distribution (NSD) data measured at an urban background site (North Kensington) in London, UK for the whole of 2011 and 2012. The PMF analyses revealed six and four factors respectively which described seven sources or aerosol types. These included Nucleation, Traffic, Diffuse Urban, Secondary, Fuel Oil, Marine and Non-Exhaust/Crustal sources. Diffuse Urban, Secondary and Traffic sources were identified by both the chemical composition and particle number size distribution analysis, but a Nucleation source was identified only from the particle Number Size Distribution dataset. Analysis of the PM_{10} chemical composition dataset revealed Fuel Oil, Marine, Non-Exhaust Traffic/Crustal sources which were not identified from the number size distribution data. The two methods appear to be complementary, as the analysis of the PM_{10} chemical composition data is able to distinguish components contributing largely to particle mass whereas the number particle size distribution dataset is more effective for identifying components making an appreciable contribution to particle number. Analysis was also conducted on the combined chemical composition and number size distribution dataset revealing five factors representing Diffuse Urban, Nucleation, Secondary, Aged Marine and Traffic sources. However, the combined analysis appears not to offer any additional power to discriminate sources above that of the aggregate of the two separate PMF analyses. Day-of-the-week and month-of-the-year associations of the factors proved consistent with their assignment to source categories, and bivariate polar plots which examined the wind directional and wind speed association of the different factors also proved highly consistent with their inferred sources.

1 Introduction

Airborne Particulate Matter (PM) is recognised as a major public health concern across the EU with costs estimated at € 600 bn in 2005 (Official Journal, 2008). In the UK

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alone, the annual health costs attributable to pollution by airborne PM were estimated in 2007 at between £ 8.5 bn and £ 18.6 bn (Defra, 2010). PM exposure was also estimated to reduce people's lives by on average seven to eight months, and by as much as nine years for vulnerable residents, such as those with asthma, living in pollution hotspots (Environmental Audit Committee, 2010). There is overwhelming evidence that both short-term and long-term exposure to ambient particulate matter in outdoor air is associated with mortality and morbidity (Pope and Dockery, 2006).

Source apportionment of airborne particulate matter has assumed increasing importance in recent years, driven by two underlying causes. Firstly, legislative pressure to reduce airborne concentrations of particulate matter has highlighted the need for reliable quantitative knowledge of the source apportionment of particulate matter in order to devise cost-effective abatement strategies. The use of source inventories alone is inadequate as these are limited in the components which they are able to quantify reliably but take no account of the different ground-level impacts of pollutants released at different altitudes or those altered by chemical transformations within the atmosphere. Some sources, such as wood burning, particle resuspension and cooking are very difficult to quantify. Consequently, there has been a need for the application of methods capable of source apportionment of ground level concentrations. Secondly, there has been a growing recognition that abatement of PM mass concentrations, taking no account of source, chemical composition or particle size, may not be a cost-effective approach if the health impact of particulate matter differs according to its source of emissions or physico-chemical characteristics. Consequently, a number of recent epidemiological studies have attempted to combine receptor modelling results with time series studies of health effects (e.g. Thurston et al., 2005; Mostofsky et al., 2012; Ostro et al., 2011).

Source apportionment methodology for particulate matter can use either receptor modelling methods or the combination of emissions inventories and dispersion modelling. The latter approach has major weaknesses associated especially with the inadequacy of emissions inventories referred to above. Consequently, most studies have

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been based upon receptor modelling methods, and in the main these have used multivariate statistical methods rather than the Chemical Mass Balance Model approach (Viana et al., 2008). The multivariate statistical approaches to source apportionment depend upon the fact that different particle sources have characteristic chemical profiles which undergo only modest changes during atmospheric transport from source to the receptor site. Such methods are also able to recognise the contributions of major secondary atmospheric constituents as a result of their characteristic chemical composition. This has led to such methods being widely used for the estimation of contributions to the mass of particles expressed as either PM_{10} or $PM_{2.5}$ (Viana et al., 2008; Belis et al., 2013).

In addition to having characteristic chemical profiles, air pollutant source categories are also likely to have characteristic partice size distributions which can also be utilised for source apportionment, although these have been utilised rather infrequently in comparison to multi-component chemical composition data. One of the few available studies (Harrison et al., 2011) used wide range particle size data collected on Marylebone Road, London, to apportion particulate matter to a total of ten sources, four of which arose from the adjacent major highway. That study used also as input data information: traffic flow according to vehicle type, meteorological factors and concentrations of gaseous air pollutants, but did not have available chemical composition data relating to simultaneous sampling of airborne particles. Other studies which have used number size distributions with chemical composition for source apportionment are Pey et al. (2009) and Cusack et al. (2013), working in Barcelona. Also in Barcelona, Dall'Osto et al. (2012) applied clustering techniques to number size distributions to identify potential sources. Such approaches are likely to be more effective close to particle sources, due to evolution of particle size distributions during atmospheric transport (Beddows et al., 2014).

One area of importance of source apportionment of airborne particulate matter arises from the fact that there are most probably differences in the toxicity of particles according to their chemical composition and size association, and as a conse-

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quence, particles from different sources may have a very different potency in affecting human health (Harrison and Yin, 2000; Kelly and Fussell, 2012). There have been many health effects studies, of which a number recently have incorporated receptor modelling methods and have sought to differentiate between the effects of different source categories on human health. Most have provided some positive and often statistically significant associations with given source factors, chemical components or size fractions (Thurston et al., 2005; Mostofsky et al., 2012; Ostro et al., 2011) but to date there is no coherence between the results of different studies and there is no generally agreed ranking in the toxicity of particles from different sources (WHO, 2013). Consequently, in this context, source apportionment methodology is tending to run ahead of epidemiology and is providing the tools for source apportionment which thus far epidemiological research has yet to utilise fully. Nonetheless, work needs to continue towards embedding source apportionment studies in epidemiological research so as to provide clearer knowledge on the toxicity of particles from different sources, or with differing chemical composition and size association.

Perhaps the most substantial variations in airborne particle properties relate to their size association, which covers many orders of magnitude. In this context, it is perhaps surprising that toxicity (expressed as effect per interquartile concentration range) appears to be of a broadly comparable magnitude for PM_{10} mass, which is determined largely by accumulation mode and coarse mode particles, and particle number which reflects mainly nucleation mode particles. Some studies, however, have suggested different health outcomes associated with the different particle metrics (e.g. Atkinson et al., 2010).

In this study, we have applied receptor modelling methods to simultaneously collected chemical composition and particle number size distribution data from a background site within central London (North Kensington). Our study has initially analysed the chemical composition and particle number size distribution datasets separately followed by analysis of the combined dataset to test whether this provides advantages in terms of greater capacity to distinguish between source categories.

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2 Experimental

2.1 Sampling site

The London *North Kensington* Site (LAT = 51.52105 and LONG = −0.213492), is part of both the London Air Quality Network and the national Automatic Urban and Rural Network and is owned and part-funded by the Royal Borough of Kensington and Chelsea. The facility is located within a self contained cabin within the grounds of Sion Manning School. The nearest road, St. Charles Square, is a quiet residential street approximately 5 m from the monitoring site and the surrounding area is mainly residential. The nearest heavily trafficked roads are the B450 (~ 100 m East) and the very busy A40 (~ 400 m South). For a detailed overview of the air pollution climate at North Kensington, the reader is referred to Bigi and Harrison (2010).

2.2 Data

For this study, 24 h air samples were taken over a two year period (2011 and 2012) using a Thermo Partisol 2025 sampler fitted with a PM_{10} size selective inlet. These were analysed for numerous chemical components listed in Table 1. For total metals (prefixed by the letter T:- Al, Ba, Ca, Cd, Cr, Cu, Fe, K, Mg, Mo, Na, Ni, Pb, Sn, Sb, Sr, V, and Zn) the concentration measured using a Perkin Elmer/Sciex ELAN 6100DRC following HF acid digestion of GN-4 Metricel membrane filters is reported. Similarly, all water soluble ions (prefixed by the letter W:- Ca^{2+}, Mg^{2+}, K, NH_4^+, Cl^-, NO_3^- and SO_4^{2-}) were measured using a near-real-time URG – 9000B Ambient Ion Monitor (URG Corp); where data from the URG was not available laboratory based ion chromatography measurements on filters (Tissuquartz™ 2500 QAT-UP) from a Partisol 2025 were used. Data capture over the two years ranged from 48 to 100 % as different sampling instruments varied in reliability. The lowest coverage was for WK (48 %), WCA (53 %), WCL (68 %), WMG (52 %) and WNH4 (50 %). All missing data was replaced using a value derived using the method of Polissar et al. (1998). A woodsmoke metric, CWOD, was

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a $PM_{10}^{Woodsmoke}$ component also included which was derived from the methodology of Sandradewi et al. (2008) utilising Aethalometer and EC/OC data, as described in Fuller et al. (2014). Concentrations of elemental carbon (EC) and organic carbon (OC) were measured by collection on quartz filters (Tissuquartz™ 2500 QAT-UP) and analysed on a Sunset Laboratory thermal-optical analyser using the QUARTZ protocol (which gives results very similar to EUSAAR 2) (NPL, 2013). Alongside the composition measurements, Number Size Distribution (NSD) data were collected using a Scanning Mobility Particle Sizer (SMPS) consisting of a CPC (TSI model 3775) combined with an electrostatic classifier (TSI model 3080) in air dried according to the EUSAAR protocol (Wiedensohler et al., 2012). The data capture of NSD over the two years was 72.5 %. Particle mass was determined on samples collected on Teflon-coated glass fibre filters (TX40HI20WW).

2.3 Positive matrix factorisation

Positive Matrix Factorisation (PMF) is a well-established multivariate data analysis method used in the field of aerosol science. PMF can be described as a least-squares formulation of factor analysis developed by Paatero (Paatero and Tapper, 1994). It assumes that the ambient aerosol X (represented by a matrix of $n \times$ observations and $m \times PM_{10}$ constituents or NSD size bins), measured at one or more sites can be explained by the product of a source matrix **F** and contribution matrix **G** whose elements are given by Eq. (1). The residuals are accounted for in matrix **E** and the two matrices **G** and **F** are obtained by an iterative minimization algorithm.

$$x_{ij} = \sum_{h=1}^{p} g_{ij} \cdot f_{hj} + e_{ij} \quad (1)$$

It is commonly understood that PMF is a descriptive model and there is no objective criterion upon which to choose the best solution (Paatero et al., 2002). This work is no exception and the number of factors and settings for the data sets were chosen using

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metrics used by Lee et al. (1999) and Ogulei et al. (2006a, b). A detailed description of PMF and our analysis is provided in the Supplement.

3 Results

The final PMF solutions were selected as those with most physically meaningful profiles. Once the PMF output is chosen and scaled, the values of the **F** matrix are used to characterise the source term. Each row i of **F** represents a source and each element f_{hj} shows the "Weight within the factor" (WWTF) of the constituent (grey bars and black NSD lines in Figs. 1–3). Together with the dimensionless **F** matrices, a parameter due to Paatero, called the Explained Variation Matrices **EV**, shows how much of the variance in the original dataset is accounted for by each factor (again see the Supplement for more details). For a given column (PM component measurement or particle size bin) of the total **EV** matrix, the Total EV (TEV) is recommended to be 0.75 or greater. Although a useful metric in assessing the ability of the final PMF settings to model the data, the **EV** values (red bars or NSD line in Figs. 1 and 2) of each factor show which constituents are the most important in each factor and hence significantly aid factor characterisation when considered alongside the WWTF. The $\mathbf{G}_i$ matrix gives the contribution of the source terms $\mathbf{F}_i$ and carries the original units of X. The values within the columns of matrix **G** contain the hourly/daily measurements made by the p factors (or sources) and are used to calculate the diurnal, weekly and yearly averages (see Figs. 1–3). The identity of the source, namely: Marine; Secondary; Traffic; Nucleation; etc., was assigned on the basis of post hoc comparison with known source profiles, tracers (Viana et al., 2008), physical properties and temporal behaviour.

3.1 The six factor solution for PM_{10} chemical composition data

An optimum six factor solution was derived which best represented the aerosol types. Figure 1 characterises the six factors as: Diffuse Urban; Marine; Secondary; Non-

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Exhaust Traffic/Crustal; Fuel Oil and Traffic. While most of the names of these factors are self-explanatory, "Diffuse Urban" needs further explanation. Diffuse Urban has a chemical profile indicative of contributions mainly from both woodsmoke (CWOD) and road traffic (Ba, Cu, Fe, Zn). Since these are ground-level sources which are affected in a similar way by meteorology (see polar plots for the Diffuse Urban and Traffic Factors in Figs. 4 and 5), PMF is not able to effect a clean separation in 24 h samples and this problem is exacerbated by the tendency of the aethalometer – which was used to derive the woodsmoke-associated CWOD variable – to include some traffic-generated carbon in the woodsmoke estimate (Harrison et al., 2013a). In the ClearfLo winter campaign, Black Carbon (traffic) from aethalometer measurements correlated strongly with the wood smoke tracer levoglucosan at North Kensington ($r^2 = 0.80$) (Crilley et al., 2015). When comparing 5, 6 and 7 factor solutions, common sources could be identified in all three solutions, namely: Diffuse Urban; Marine; Secondary; Non-Exhaust Traffic/Crustal; and Fuel Oil. In the 5 factor solution, the Diffuse Urban factor had elevated values of EC, Ba, Cu, Fe, Mg, Mn and Sb all of which are indicative of a traffic contribution. By increasing the number of factors from 5 to 6, the concentration of these elements within the Diffuse Urban factor decreased as a Traffic factor separated out into its own unique factor, although a complete separation was not observed even when using 7 factors. Furthermore, when using 7 and 8 factors, the Diffuse Urban factor remained unaltered and the Fuel Oil factor was observed to shed a spurious factor containing odd combinations of nickel, lead, zinc, sulphate, and organic carbon contributions. This led to the conclusion that only 6 factors yielded a meaningful solution.

Considering further the 6 factor solution, the Marine factor clearly explains much of the variation in the data for Na, Cl^- and Mg and the Secondary factor is identified from a strong association with NH_4^+, NO_3^-, SO_4^{2-} and organic carbon. Considering traffic emissions, the PM does not simply reflect tailpipe emissions, but also includes contributions from non-exhaust sources, including the re-suspension of road dust and primary PM emissions from brake, clutch and tyre wear (Thorpe and Harrison, 2008). The Non-Exhaust Traffic/Crustal factor explains a high proportion of the variation in the

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Al, Ca and Ti measurements consistent with particles derived from crustal material; derived either from wind blown or vehicle induced resuspension. There is also a significant explanation of the variation in elements such as Zn, Pb, Mn, Fe, Cu and Ba which have a strong association with non-exhaust traffic sources. As there is a strong contribution of crustal material to particles resuspended from traffic (Harrison et al., 2012), it seems likely that this factor is reflecting the presence of particulate matter from resuspension and traffic-polluted soils.

The fifth factor, attributed to Fuel Oil, is characterised by a strong association with V and Ni together with significant SO_4^{2-}. These are all constituents typically associated with emissions from fuel oil combustion. The sixth factor shows an especially strong association with elements derived from brake wear (Ba, Cu, Mo, Sb) and tyre wear (Zn) (Thorpe and Harrison, 2008; Harrison et al., 2012). There is also a strong association with elemental carbon (EC) and organic carbon (OC) in a ratio of approximately 2 : 1 consistent with exhaust emissions from road traffic resulting from factor pulling the EC and OC ratios. This factor is therefore assigned the title of Traffic.

Also shown in Fig. 1 is a pie chart showing the proportion of mass concentration associated with each of the factors and bar charts showing the day-of-the-week dependence and monthly dependence of the average concentration associated with each factor. Three sources predominate: non-exhaust and crustal (25 %), secondary (25 %) and diffuse urban (24 %), with lesser contributions from marine (15 %), local traffic (5 %) and fuel oil (6 %). Both the Traffic and Non-Exhaust Traffic/Crustal factors show higher concentrations on weekdays than at weekends reflecting traffic activity in London. The Diffuse Urban source shows slightly higher concentrations at weekends likely to be a reflection of recreational wood burning (Fuller et al., 2014). The Marine and Fuel Oil factors show no consistent variation with day-of-the-week. In the case of the monthly variations, the Diffuse Urban, Marine, Secondary and Non-Exhaust Traffic/Crustal sources all show signs of higher concentrations in the cooler months of the year. Both the Diffuse Urban and traffic-related sources are emitted at ground-level and are likely to be less well dispersed in a shallower mixing layer during the colder

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months of the year. Marine aerosol typically shows a seasonal variation with elevated concentrations associated with the stronger winds in the winter months. The secondary constituent is particularly strong in the spring which is when nitrate concentrations are typically elevated (Harrison and Yin, 2008), probably as a result of relatively low air temperatures suppressing the dissociation of ammonium nitrate and increased emissions of ammonia due to the spreading of slurry on farmland. The only constituent to show higher concentrations in the warmer months of the year is the Fuel Oil source. This might be attributable to emission from high chimneys with more efficient mixing to ground level during the more convective summer months.

Figure 6 plots how the factors contributed daily across the 2 year data set to the total measured PM_{10}, and the vertical dotted lines identify the period containing the highest contribution of each factor to the PM_{10} mass concentration. Air mass back trajectories corresponding to these periods have been calculated using HYSPLIT (Draxler and Rolph, 2015) and are shown in Fig. 7. As expected, the largest contribution of the Marine factor was associated with a long (i.e. high average wind-speed) maritime trajectory associated with marine aerosol production. The Secondary factor was associated with winds from the European mainland crossing the Benelux countries *en route* to the North Kensington site. This trajectory sector from London was identified by Abdalmogith and Harrison (2005) as strongly associated with elevated sulphate and nitrate concentrations.

The Traffic factor was associated with a trajectory travelling across eastern and northern France before crossing the English Channel to the UK, approaching the North Kensington site from the south-east. Such a trajectory is likely to maximise both the long-range advected contribution and the local contribution within London. The highest contribution from the Diffuse Urban factor was during the identical period to the highest traffic contribution and hence the identical back trajectories. Examination of Fig. 6 shows many similar features in the time series of the Diffuse Urban and Traffic source categories which confirm the impression that road traffic makes a substantial contribution to the Diffuse Urban factor. The maximum contribution from the Non-

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Exhaust/Crustal factor was again on an easterly circulation rather similar to that giving a maximum in the Secondary contribution (Fig. 7). This trajectory was likely to include a substantial contribution from air advected from mainland Europe but also in air from the centre and east of London. Perhaps most interesting is the trajectory associated with the highest contribution of the Fuel Oil factor which shows air arriving predominantly from the English Channel and remaining at low altitude confirming the impression that there may be a major contribution from shipping to the Fuel Oil factor. This would be consistent with the observation of Johnson et al. (2014) that shipping was the main source impacting upon V in Brisbane, Australia and that this was associated with both sulphur and black carbon. In our data shown in Fig. 1, the fuel oil factor accounted for almost 75 % of the explained variation of V. Receptor modelling of airborne PM collected in Paris (France) revealed a Heavy Oil Combustion source which accounted for a high percentage of V and Ni, and some SO_4^{2-}, with a predominant source area around the English Channel (Bressi et al., 2014), consistent with a substantial influence of shipping emissions.

Table 2 shows the average concentrations of gas phase pollutants and meteorological conditions corresponding to the period when each factor in the PMF results for PM_{10} chemical composition exceeded its 90 percentile value. Notable amongst these are the high carbon monoxide and NO_x concentrations associated with the Traffic and Diffuse Urban sources and the relatively clean air associated with the Marine source.

3.2 The four factor solution for the Number Size Distribution (NSD) data

The PMF analysis of the hourly averaged measurements collected at North Kensington (2011–2012) yielded an optimum four factor solution. Figure 2 characterises the four factors as: Secondary, Diffuse Urban, Traffic and Nucleation. Comparing this optimum solution with its counterparts using 3 and 5 factors, all three solutions had in common a Traffic and Diffuse Urban factor. Using 3 factors, the Nucleation and Secondary factors were combined and only separated when using 4 factors. When using 5 factors, the Secondary factor divided again, shedding an obscure factor with three modes at

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~ 0.03, ~ 0.08 and $\sim 0.3\,\mu m$, all equally spaced along the $\log_{10}(D_a)$ axis. This spurious factor had a noticeable correlation with its parent factor suggesting factor splitting at 5 factors leading to a conclusion that only 4 factors could be used to obtain a meaningful solution. Figure 2 also shows the weekday/weekend and seasonal behaviour of these factors, the number size distributions associated with each factor together with the explained variation for each size bin within each factor. The right-hand panels show the diurnal variation of each factor and the variance explained for each time-of-day. Figure 8 plots how these factors contributed on a daily basis across the two year dataset to the total NSD measured.

The Secondary factor shows by far the coarsest particle sizes with a minimum concentration in the early afternoon likely associated with the evaporation of ammonium nitrate at higher air temperatures and relative humidities. There is no consistent day-of-the-week pattern and elevated concentrations in spring presumably arise for the same reasons as for the PM_{10} Secondary constituent. The Traffic factor has a modal diameter at around 30 nm and a large proportion of the variation explained within the main peak of the distribution. The diurnal pattern has peaks associated with the morning and evening rush hour periods and there are lower concentrations at weekends and higher concentrations in the winter months of the year. All of these features are consistent with emissions from road traffic (Harrison et al., 2011). The factor described as Diffuse Urban has a modal diameter intermediate between that of the Traffic and Secondary factors and a diurnal pattern consistent with that expected for traffic emissions. Its concentrations are elevated at weekends, presumably associated with recreational wood burning and higher concentrations in the cooler months of the year. The fourth factor which is attributed to Nucleation has by far the smallest particle mode at around 20 nm and peaks around 12 noon in association with peak solar intensities. It shows a seasonal cycle with the highest concentrations on average in the summer months in year 2 (Fig. 8) and a preference for weekday over weekend periods. The apparent lack of a seasonal pattern in the first year of observations is surprising. However, nucleation depends upon a complex range of variables including precursor availability, insolation

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and condensation sink, and the reasons are unclear. The apparent background level of nucleation in year 2 accounting for up to 1000 cm^{-3} particles may be the result of an incomplete separation of this factor from other source-related factors.

The mean particle number concentration, measured using the SMPS was 5512 cm^{-3}, of which Traffic and Diffuse Urban made the highest percentage contribution of 44.8 and 43.0 % respectively, followed by Nucleation (7.8 %) and Secondary (4.4 %).

Figure 8 includes dotted vertical lines which identify the days with the highest average contribution of each factor to the total particle number concentration and the air mass back trajectories corresponding to these periods have been plotted in Fig. 9. This shows some differences relative to the factors derived from the PM_{10} composition dataset. The Secondary factor trajectories originated over the North Sea and the majority crossed parts of Germany and the Netherlands, on a more northerly path than the trajectories of the PM_{10} Secondary factors. The trajectory for the Diffuse Urban source had crossed over North Eastern France before arriving at NK in a similar manner to the PM_{10} Diffuse Urban trajectory. The Traffic factor back trajectory approached from the west after crossing the southern UK which is quite different to the PM_{10} Traffic factor seen in Fig. 7 and the Nucleation factor was associated with relative low wind speeds and crossing the southern UK before reaching the sampling site. The Nucleation factor is predominantly maritime and therefore likely to bear a rather low aerosol concentration hence favouring the nucleation process. Table 2 presents the average gas phase pollutant concentrations and meteorological conditions corresponding to the peak contribution of the various factors. Notable amongst these are the low concentrations of carbon monoxide, oxides of nitrogen, sulphur dioxide and high ozone concentration associated with the Nucleation factor.

In Table 3 the correlation coefficients are given between the factors derived from the PM_{10} composition dataset and those from the NSD dataset. There are moderate correlations between the Diffuse Urban factors determined from the two PMF analyses and for the Secondary factors. The Traffic factor in the PM_{10} dataset has a higher

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correlation with the Diffuse Urban factor derived from the NSD dataset than with the Traffic factor from that dataset, and the Diffuse Urban factor from the PM_{10} dataset shows a very modest correlation with the Traffic factor from the NSD dataset. This serves to confirm the contribution of traffic to the Diffuse Urban factor. The Nucleation factor in the NSD dataset and Marine and Fuel Oil factors in the PM_{10} composition dataset do not correlate substantially with factors in the other dataset.

3.3 Combined PM_{10} and NSD data

The PM_{10} composition and daily average NSD datasets were combined into one daily PM_{10}_NSD data set and analysed using PMF2. By combining the two datasets, an apportionment was made that was sensitive to both particle number and mass composition of the sources. This resulted in a five factor solution which was described by the factors interpreted as: Diffuse Urban, Nucleation; Secondary; Marine and Traffic (Fig. 3). The factor with the smallest mode in the number size distribution (around 25 nm) was attributed to Nucleation. It showed chemical association with species such as sulphate, nitrate, ammonium and organic carbon (OC) and had a slight preference for weekdays over weekends (Fig. 3) and a strong association with the summer months of the year. There is also a well defined traffic factor which has a mode at around 30 nm as observed previously for road traffic (Harrison et al., 2012) as well as chemical associations with Al, Ba, Ca, Cu, Fe, Mn, Pb, Sb, Ti and Zn. This factor clearly therefore encompasses both the exhaust and non-exhaust emissions of particles. A factor which can be clearly assigned on the basis of its chemical association is that described as Aged Marine. This explains a large proportion of the variation in Na, Mg and Cl but shows a number size distribution with many features similar to that of the Traffic factor with which it has rather little in common chemically. Since the aged marine mass mode is expected to be in the super-micrometre region and hence well beyond that measured in the NSD dataset, it seems likely that the size distribution associated is simply a reflection of other sources influencing air masses rich in marine particles.

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The Secondary factor is assigned largely on the basis of strong associations with nitrate, sulphate, ammonium and organic carbon (OC). The number size distribution shows a mode at around 85 nm and a mode is also seen in the volume size distribution at 0.3–0.4 µm. The Diffuse Urban factor has chemical associations with non-exhaust traffic sources (Ba, Cu, Fe, Mo, Pb, Sb, Zn) as well as exhaust emissions (elemental carbon (EC), organic carbon (OC)) and the woodsmoke indicator (CWOD). The particle size mode at around 55 nm is coarser than anticipated for traffic emissions and appears to be strongly influenced by emissions of woodsmoke. This factor, along with the Secondary factor, shows a predominance of weekend over weekday abundance (Fig. 3), whereas the Nucleation and Traffic factors show a greater association with weekdays than weekends. Also seen in Fig. 3, the Nucleation factor has an enhanced abundance in the summer months while the Diffuse Urban and Traffic factors are more abundant in the cooler months of the year. As in the PM_{10} mass composition and Number Size Distribution analyses, the Secondary factor shows a dominance of concentrations measured in the Spring, presumably reflecting the well reported elevation in nitrate concentrations in the UK at that time of year (Harrison and Yin, 2008).

3.4 Polar plots

Figure 10 shows bivariate polar plots for the PMF factors derived from the combined chemical composition/NSD analysis which describe the wind direction (angle) and wind speed (distance from centre of plot) dependence of the factors using the Openair project software (Carslaw and Ropkins, 2012). The Diffuse Urban factor has an association with all wind directions and a predominant occurrence at low wind speeds. There is also a stronger association with easterly winds than with other wind directions and here it was present at higher wind speeds. This is consistent with the North Kensington site being in the west of central London and therefore both the greatest density of London sources and the influence of pollutants advected from the European mainland are associated with easterly winds. Broadly similar behaviour is seen for the Traffic factor with an association with low wind speeds and easterly wind direction, again most

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probably reflecting the higher density of sources in this wind sector, and possibly also the greater tendency for low wind speeds associated with easterly circulations which are frequently anticyclonic. The Secondary source also shows a strong association with easterly winds and a predominant association with moderate wind speeds which is known to be associated with secondary pollutants in easterly air masses frequently advected from the European mainland (Abdalmogith and Harrison, 2005). The plots for both Nucleation and Aged Marine factors are very different from the Diffuse Urban, Secondary and Traffic sources, and show distinct differences from one another. The Nucleation factor is associated primarily with moderate wind velocities in the west south-westerly sector. This is a sector most often associated with relatively clean Atlantic air which most probably favours the nucleation process due to the low condensation sink in air masses with a lower aerosol surface area. On the other hand, the aged marine factor is associated primarily with south-westerly winds of high strength reflecting the requirement for maritime air and high wind speeds. There is also some association with other wind sectors due to the presence of seas all around the United Kingdom, but in all cases there is a requirement for high wind speeds to generate the marine aerosol.

Figure 4 presents the bivariate polar plots for the output of the PMF run on the PM_{10} mass composition data. The plots for the Diffuse Urban, Marine, Secondary and Traffic factors are very similar to those seen in Fig. 10. The PMF on mass composition data is unable to identify a Nucleation factor but identifies separate Non-Exhaust/Crustal and Fuel Oil factors. The polar plot for the Non-Exhaust and Crustal factor shows slightly more northerly wind direction dependence than for the Traffic factor and an appreciably higher dependence on wind speed. This is strongly suggestive of a wind-driven resuspension contribution to this factor, but the association with more easterly winds as for the Traffic factor in Fig. 10 indicates association with road traffic. The Fuel Oil factor seen in Fig. 4 is quite different, with the polar plots suggesting a range of sources in the sector between east and south of the sampling site and associations with a wide range of wind speeds including relatively strong winds. This may be an indication of

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a contribution of emissions from shipping burning fuel oil while travelling through the English Channel to the south-east of London. The major difference from all other polar plots confirms this as a highly distinctive source category.

Figure 5 shows both bivariate polar plots (wind direction and wind speed in the left-hand panels) and annular plots showing both wind direction and time-of-day in the right-hand panels for the output for the PMF analysis of the Number Size Distribution data. The Nucleation factor has a very clear behaviour with predominant associations with westerly winds and occurrence in the afternoon when particles have grown sufficiently in size to cross the lower size threshold of the SMPS instrument used. In this case, however, some association with winds from a variety of direction sectors is seen unlike Fig. 10. The Traffic factor again shows a predominant association with easterly winds, although there is some clear association with light westerly winds also. The predominant temporal association is with the morning rush hour and late evening, consistent with the lower temperatures and restricted vertical mixing typical of such times of day combined with high levels of traffic emissions. The Diffuse Urban source, as in Fig. 10, has a predominant association with the easterly wind sector, and there is also a clear temporal association with the morning rush hour and the late evening reflecting both traffic emissions (as for the Traffic factor) and most probably also wood burning emissions in the evening data. The final Secondary factor shows an association with winds from northerly through to south-easterly and a predominance of the cooler hours of the day favouring the presence of semi-volatile ammonium nitrate in the condensed phase. Overall, these plots and those for the PM_{10} mass composition data are highly consistent with those from the combined PM_{10} mass composition/Number Size Distribution data analysis.

4 Discussion

This work gives quantitative insights into the sources of airborne particulate matter at a representative background site in central London averaged over a two year period.

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The results for PM mass complement recent work on $PM_{2.5}$ mass which compared the implementation of a Chemical Mass Balance model using organic and inorganic markers with source attribution by application of PMF to continuous measurements of non-refractory chemical components of particulate matter using an Aerosol Mass Spectometer (AMS) (Yin et al., 2015) and also the AMS PMF carried out by Young et al. (2014). It must be remembered that the AMS is also limited to sampling non-refractory aerosol and $PM_{0.8}$ which will be different to the composition of PM_{10} considered in this study. The lack of full resolution of the ground-level combustion source contribution in the current study is disappointing, and while the complementary CMB (Yin et al., 2015) and AMS (Young et al., 2014) work gives additional valuable insights, neither quantifies the contribution to the PM_{10} size fraction addressed in this study, and the labour-intensive CMB work covers a period of only one month.

The present method based upon multi-component analysis and the application of PMF is less intensive in terms of data collection than the CMB model approach, but when applied to urban air it is a relatively blunt tool. In common with other urban studies, it is able to identify about six separate source categories (Belis et al., 2013) but there is inevitably some question of how cleanly these have been separated and what sub-categories may have contributed to the data but failed to be recognised. This study could not make a clean separation of the the Diffuse Urban from wood burning and traffic factors which will tend to show a broadly similar day-to-day variation as they are both very widespread ground level sources affected in a similar way by meteorology, and thus strongly correlated. To achieve a separation of the sources would probably require the analysis of levoglucosan as a highly selective tracer for biomass combustion. A further factor which was identified by both CMB modelling and by AMS (Yin et al., 2015) is emissions from food cooking which increasingly are seen as a significant contributor to particulate matter in urban atmospheres. This is a component which can vary significantly in composition according to the specific source and hence presents considerable challenges for quantification. There is no specific or highly selective tracer for

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cooking (other than cholesterol for meat cooking). With the absence of a cooking tracer within this study, this source most probably resides within the Diffuse Urban factor.

While, because of different sampling periods, a quantitative comparison of the results of this study with those obtained by Yin et al. (2015) in a CMB study of the North Kensington site in London is of very limited value, it is worthwile to compare the source categories identified. One of the major differences between the PMF and CMB methods is that CMB requires prior knowledge of source categories and corresponding chemical profiles whilst PMF makes no a priori assumptions on these points. The CMB model (Yin et al., 2015) used as input source categories, vegetative detritus, wood smoke, natural gas, dust/soil, coal, food cooking, traffic, biogenic secondary, other secondary, sea salt, ammonium sulphate and ammonium nitrate. Of those, there is direct overlap between the PMF Marine and CMB Sea Salt categories and the PMF Secondary factor and the CMB ammonium sulphate/nitrate classes. The CMB model estimates traffic on the basis of organic molecular markers which respond particularly to the exhaust emissions, while it is clear that both the PMF factors for Traffic and Non-Exhaust Traffic/Crustal both contained a substantial contribution from non-exhaust sources. The Diffuse Urban factor in the PMF modelling probably has a strong overlap with the woodsmoke and a proportion of the traffic contribution estimated by the CMB model. The vegetative detritus, natural gas, coal and food cooking sources contributed by the CMB model were not differentiated by the PMF and probably appear largely within the Diffuse Urban category. On the other hand, the Fuel Oil factor, which emerges very clearly from the PMF analysis, was not apparent in the CMB results which did not use suitable chemical tracers. Consequently, the two methods appear to be largely complementary, and there could be benefits in use of the PMF method to inform the source categories used in the CMB modelling.

There is a question of whether there was any advantage in combining mass composition data and number size distribution data in the source apportionment calculations. As anticipated, the data analyses based upon chemical composition alone and upon particle number size distributions alone were able to elucidate many components in

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common, but also some which were unique to each method. It is unsurprising that the analysis of chemical composition data was, for example, unable to elucidate a Nucleation factor which has little impact on particle mass but a substantial impact upon particle number. From a source perspective, the combination of the two datasets did not provide additional insights, and the best outcomes appeared to have arisen from analysis of the mass composition and number size distribution datasets separately with a combined view of the results. For future health studies the relative merits of focusing on particle mass or particle number will depend on the balance of emerging information on which metric is most closely associated with human health effects, or whether each metric is associated with different health outcomes.

The pie chart in Fig. 1 indicates that substantial reductions in PM_{10} mass could be achieved by abatement of the Diffuse Urban (woodsmoke, traffic and probably cooking) and traffic sources, the latter contributing to three of the factors (Traffic, Diffuse Urban and Non-exhaust Traffic/Crustal). This may prove more effective than reductions in the secondary component, for which non-linear precursor-secondary pollutant relationships challenge the effectiveness of abatement measures (Harrison et al., 2013b).

Nanoparticles (measured by the NSD distributions) contribute little to particulate mass, but might play an important role in the toxicity of airborne particulate matter with epidemiology from London showing a significant association of cardiovascular health outcomes with nanoparticle exposure (as reflected by particle number count, Atkinson et al., 2010). In our work, we saw a substantial contribution of traffic (44.8 %) to PN which contrasts with the much lower contribution (4.5 %) to PM_{10} mass. The fine fraction comes mainly from primary emission from combustion sources and from Fig. 2 we see that the Diffuse Urban factor was the second largest contributor (43.0 %) to PN followed by relatively minor impacts from Secondary and Nucleation processes (combined sum of 12.2 %). This clearly indicates that combustion contributes the majority of urban nanoparticles; consistent with road traffic emissions being recognised as the largest source of nanoparticles in the UK national emissions inventory (AQEG, 2005).

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Table 1. Measurements collected at the North Kensington Site, 2011 and 2012.

Species	Brief Description	PM Fraction	Detailed Description
TMN	Manganese	PM_{10}	Total metal concentration –
TMO	Molybdenum		HF acid digest and ICPMS
TNA	Sodium		
TNI	Nickel		
TPB	Lead		
TSB	Antimony		
TSN	Tin		
TSR	Strontium		
TTI	Titanium		
TV	Vanadium		
TZN	Zinc		
TAL	Aluminium		
TBA	Barium		
TCA	Calcium		
TCD	Cadmium		
TCR	Chromium		
TCU	Copper		
TFE	Iron		
TK	Potassium		
TMG	Magnesium		
PCNT	Particle Number	PM_1	Condensation particle counter (CPC, TSI)
PM_{10}	PM_{10}	PM_{10}	EU reference equivalent. Gravimetric with gaps filled from FDMS-TEOM
PM_{25}	$PM_{2.5}$	$PM_{2.5}$	EU reference equivalent. FDMS-TEOM with gaps from gravimetric
EC	Elemental Carbon	PM_{10}	By thermo chemical analysis using Sunset instrument and NIOSH TOT protocol.
OC	Organic Carbon	PM_{10}	
CWOD	OA Wood Burning	$PM_{2.5}$	OA from wood using uses aethalometer wood burning model of Sandradewi et al., 2008 as in Fuller et al., 2014
WNO_3	Nitrate	PM_{10}	Water souble measured using near real time URG, gaps filled with filter measurements
WSO_4	Sulphate		
WCL	Chloride		
WNH_4	Ammonium		
WCA	Calcium		
WMG	Magnesium		
WK	Potassium		

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Table 2. Average concentrations of gas phase pollutants and meteorological conditions corresponding to the periods when each factor in the PMF results for the PM_{10} chemical and NSD exceeded its 90 %ile value.

PM_{10}	CO mg m^{-3}	NO µg m^{-3}	NO_2 µg m^{-3}	NO_x µg m^{-3}	O_3 µg m^{-3}	SO_2 µg m^{-3}
Traffic	0.43	50.02	62.59	139.05	12.42	3.71
Fuel Oil	0.20	4.42	27.63	34.33	46.82	1.25
Non-Exhaust/Crustal	0.35	26.64	53.71	94.67	24.50	3.48
Secondary	0.28	18.09	48.79	76.61	48.65	3.23
Marine	0.22	5.69	29.48	38.40	46.54	2.04
Diffuse Urban	0.38	42.69	61.42	126.46	20.15	3.91
NSD	**CO mg m^{-3}**	**NO µg m^{-3}**	**NO_2µg m^{-3}**	**NO_x µg m^{-3}**	**O_3 µg m^{-3}**	**SO_2 µg m^{-3}**
Secondary	0.38	30.72	57.48	104.63	25.93	3.75
Diffuse Urban	0.39	44.19	60.43	128.19	23.84	3.58
Traffic	0.32	29.70	54.04	99.91	20.63	2.77
Nucleation	0.24	9.31	33.52	47.88	37.00	2.23

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Table 2. Continued.

PM_{10}	WD degrees	WS ms^{-1}	VIS m	*P* mBar	*T* °C	DP °C	RH %
Traffic	196	4.79	1197	1022	6.01	3.01	81.93
Fuel Oil	205	11.25	2239	1015	11.41	6.93	75.47
Non-Exhaust/Crustal	134	5.56	951	1023	9.09	5.37	79.33
Secondary	152	6.17	1687	1019	14.98	7.90	65.34
Marine	203	7.84	2085	1015	16.24	11.15	73.93
Diffuse Urban	166	4.87	1405	1020	11.33	6.64	76.54
NSD	WD degrees	WS ms^{-1}	VIS m	*P* mBar	*T* °C	DP °C	RH %
Secondary	141	5.14	878	1022	10.73	6.33	76.68
Diffuse Urban	168	4.67	1266	1021	10.64	6.13	76.63
Traffic	193	5.79	1903	1020	9.27	5.14	77.51
Nucleation	206	7.95	2103	1015	12.8	7.9	74.27

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Table 3. Pearson correlation coefficients between the daily average NSD and PM_{10} factors.

Factors			NSD 1 Secondary	2 Diffuse Urban	3 Traffic	4 Nucleation
PM_{10}	1	Diffuse Urban	0.60	0.77	0.414	−0.07
	2	Marine	−0.36	−0.35	−0.127	−0.09
	3	Secondary	0.64	0.30	−0.006	−0.15
	4	Non-Exhaust Traffic/Crustal	0.47	0.41	0.097	−0.14
	5	Fuel Oil	−0.14	0.02	−0.070	0.28
	6	Traffic	0.53	0.72	0.471	−0.08

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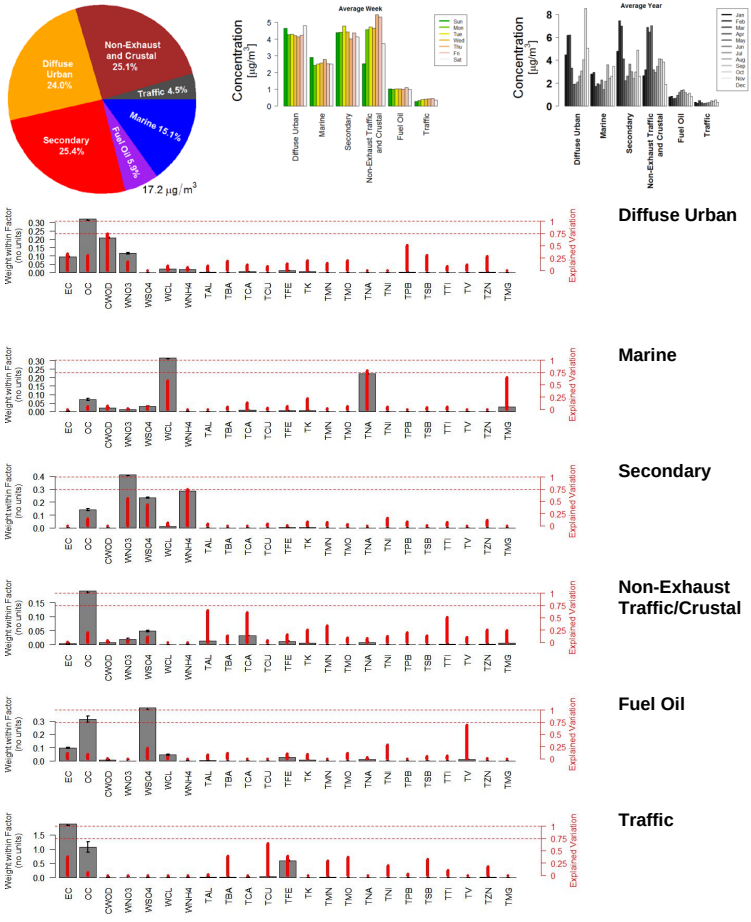

Figure 1. Factors outputted from PMF2 run on PM_{10} mass composition data showing the contribution (grey bar) and Explained Variation of each metric (red bar).

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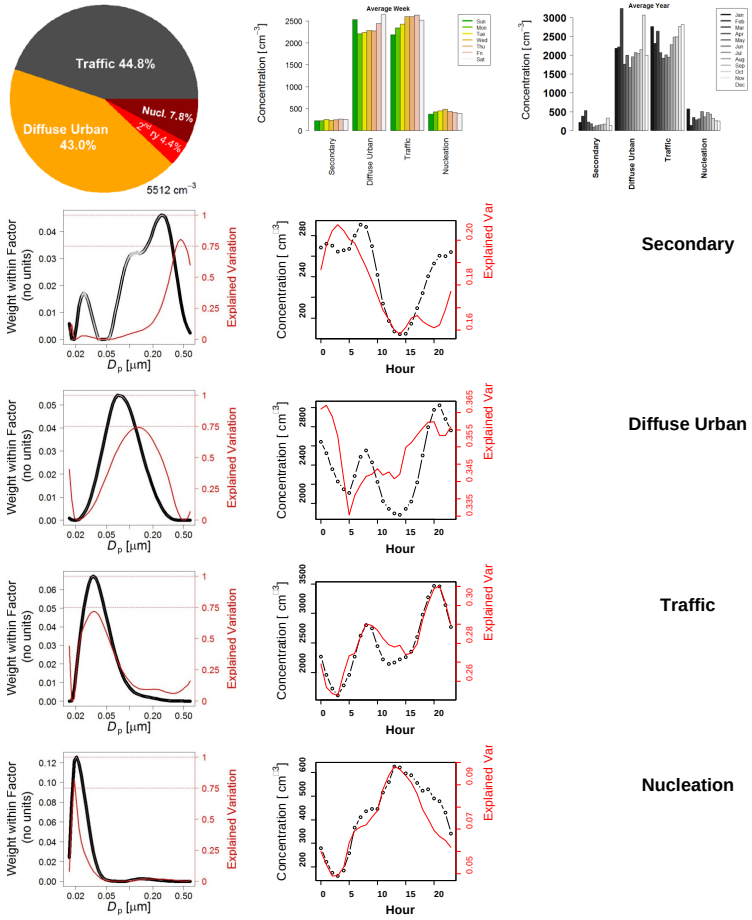

Figure 2. Factors outputted from PMF2 run on the Particle Number Size Distribution showing the contribution (black line) and Explained Variation of each metric (red line).

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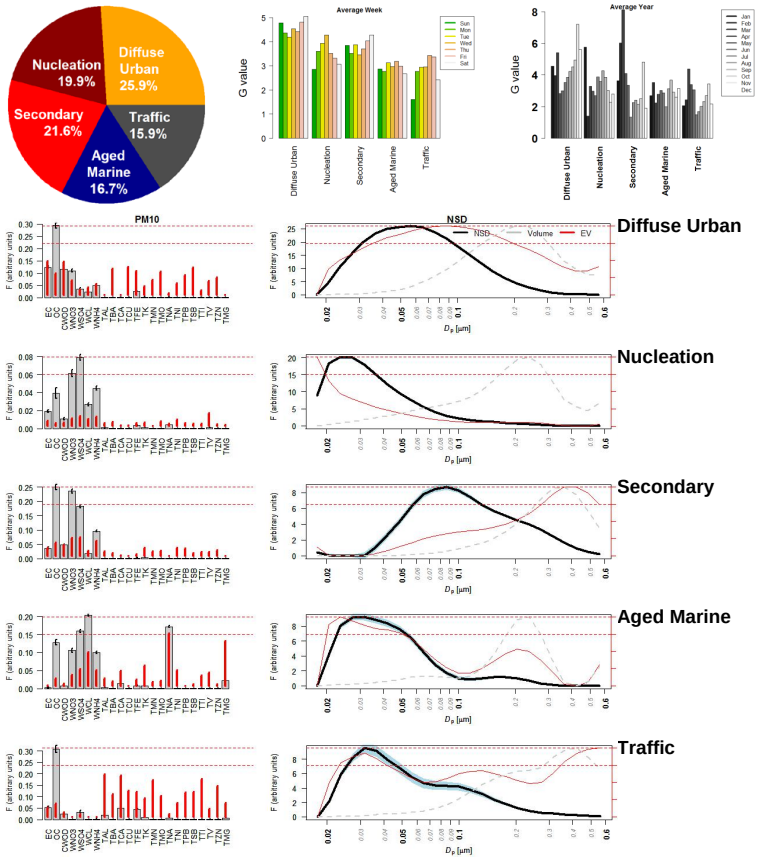

Figure 3. Five factor solution from the combined composition/NSD dataset showing the contribution (black line) and Explained Variation of each metric (red line).

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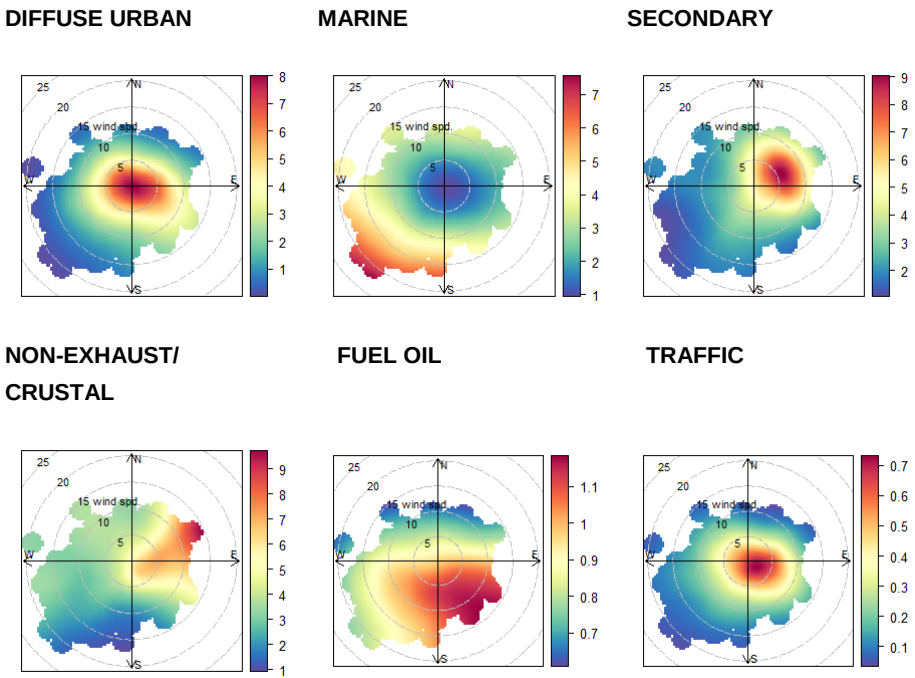

Figure 4. Polar plots showing how the daily PM_{10} contributions are affected by the daily vector average wind direction and velocity. (Units: PM_{10} ($\mu g\,m^{-3}$) and wind speed ($m\,s^{-1}$).)

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15, 10123–10162, 2015

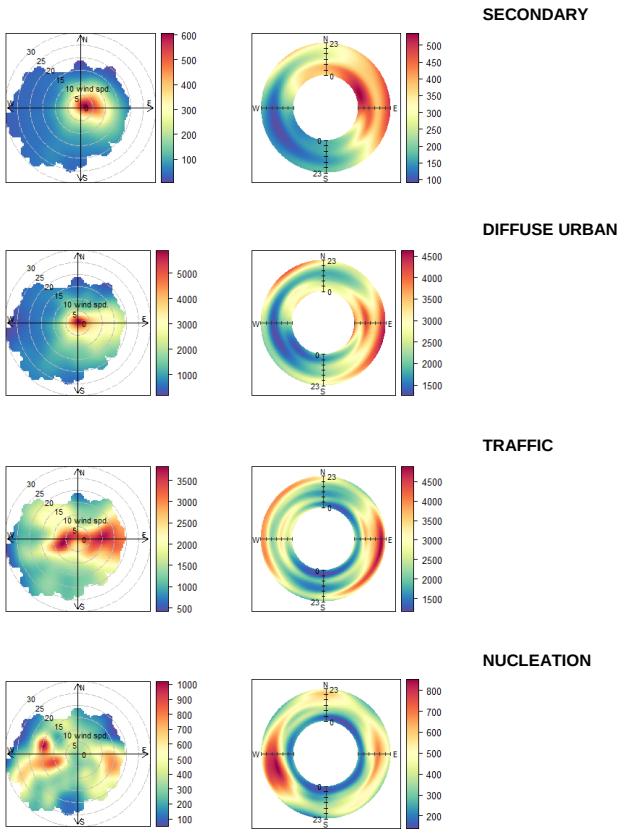

Figure 5. Polar plots showing how the hourly NSD contributions are affected by the hourly wind direction and wind velocity. (Units: NSD (cm^{-3}) and wind speed ($m\,s^{-1}$).)

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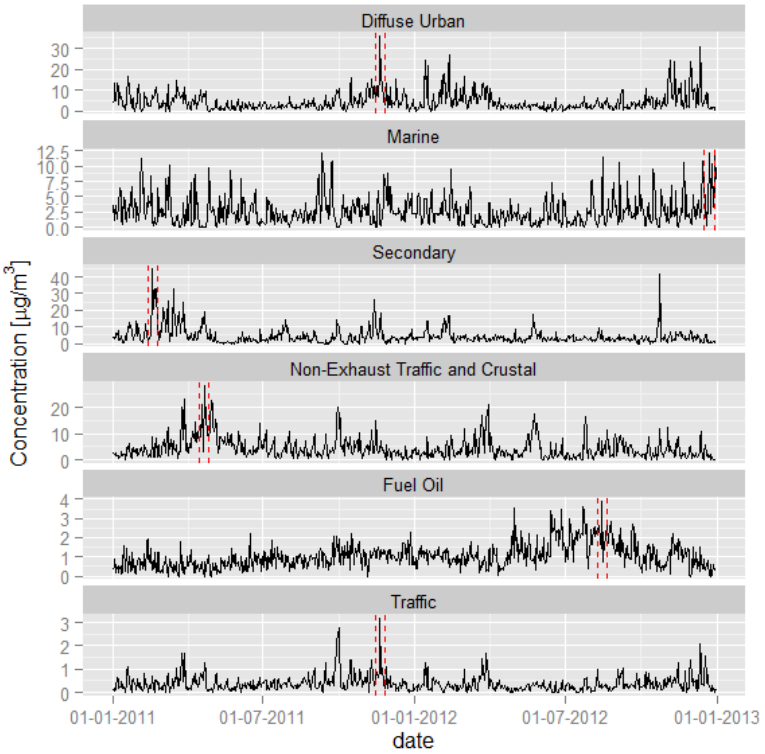

Figure 6. Daily Factor Scores outputted from PMF2 GF. (Vertical red lines indicate when each factor has the highest contribution to PM_{10}. 20 November 2011 – Diffuse Urban; 23 December 2012 – Marine; 18 February 2011 – Secondary; 21 April 2011 – Non-Exhaust and Crustal; 15 August 2012 – Fuel Oil; 20 November 2011 – Traffic.)

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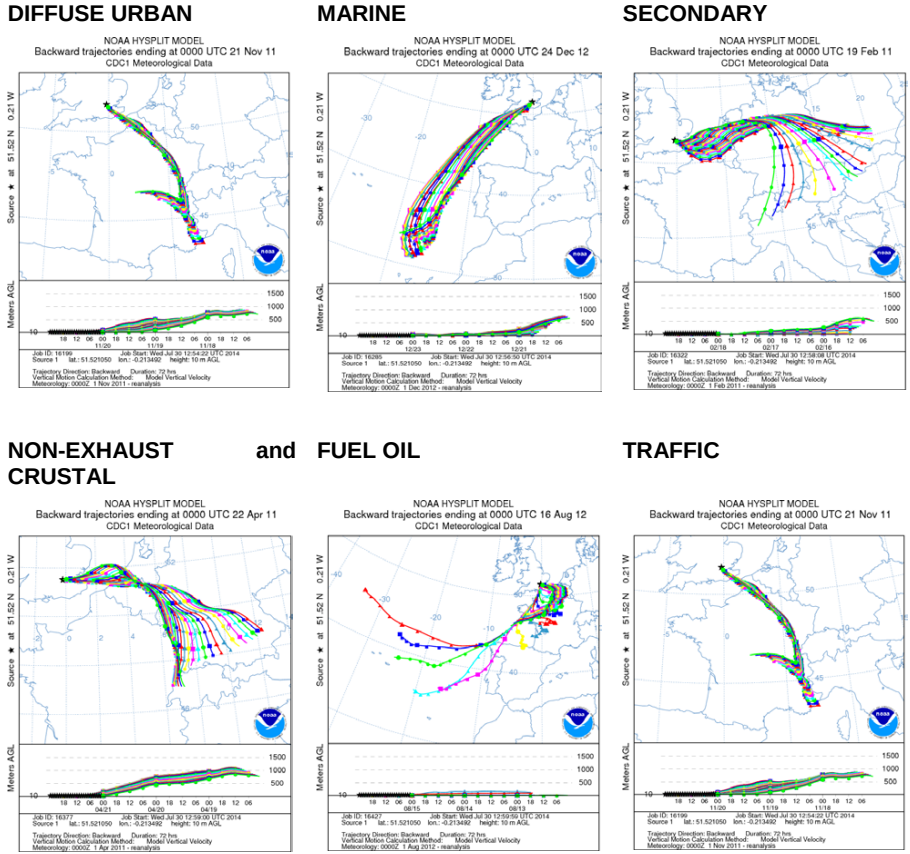

Figure 7. Back trajectories corresponding the vertical red lines in Fig. 6, which indicate when each factor has the highest contribution to PM_{10} (20 November 2011 – Diffuse Urban; 23 December 2012 – Marine; 18 February 2011 – Secondary; 21 April 2011 – Non-Exhaust and Crustal; 15 August 2012 – Fuel Oil; 20 November 2011 – Traffic).

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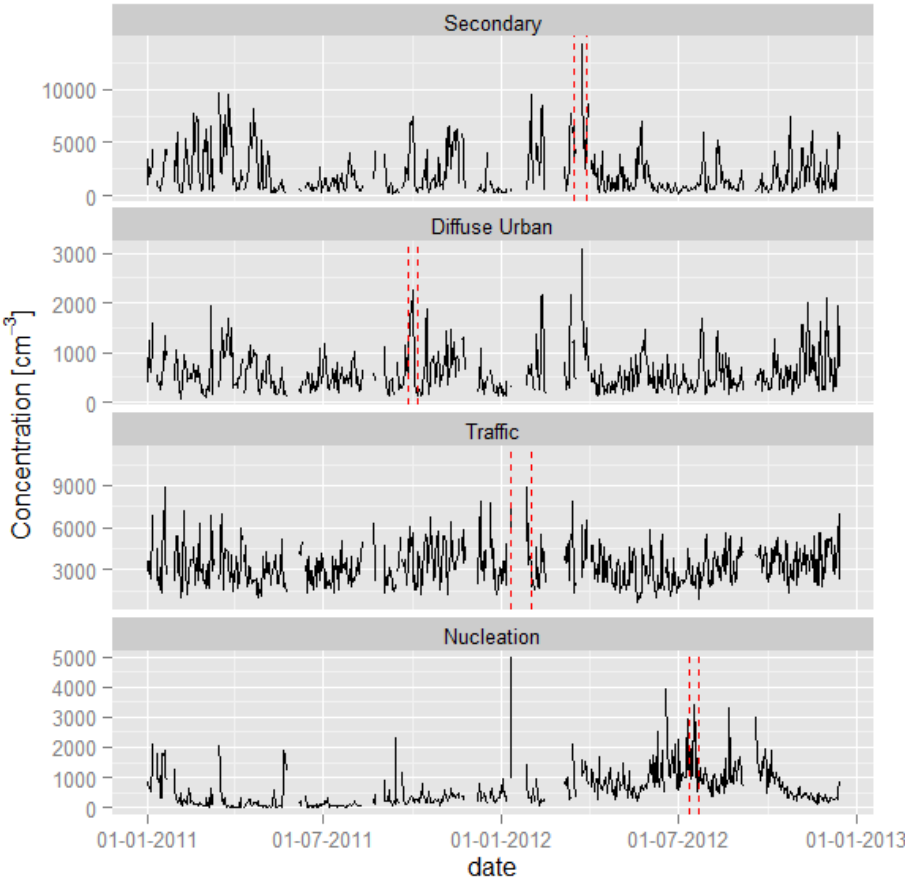

Figure 8. Daily Factor Scores outputted from PMF2 GF (unit cm^{-3}). (Vertical red lines indicate when each factor has the highest daily average contribution to the NSD. 24 March 2012 – Secondary; 1 October 2011 – Diffuse Urban; 27 January 2012 – Traffic; 17 July 2012 – Nucleation.)

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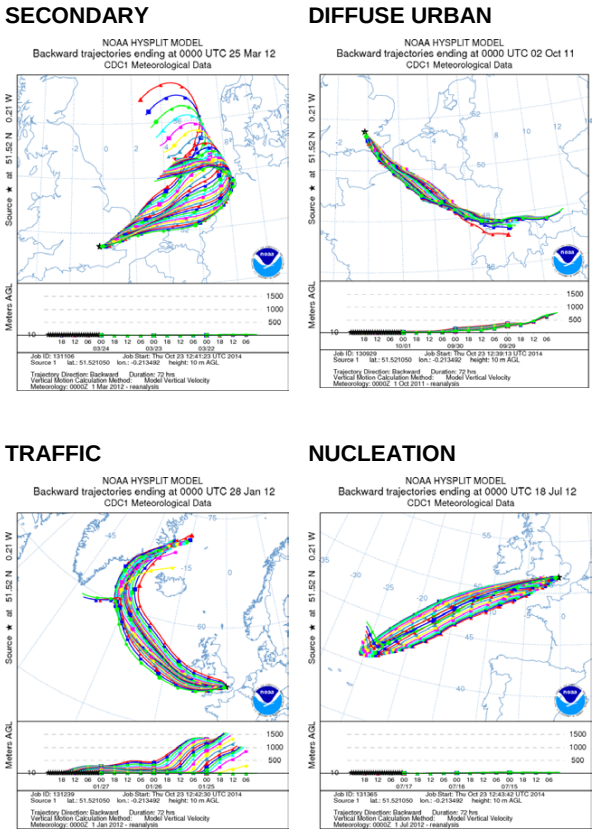

Figure 9. Back mass trajectories corresponding the vertical red lines in Fig. 8, which indicate the day each factor has the highest daily contribution to NSD. (24 March 2012 – Secondary; 1 October 2011 – Diffuse Urban; 27 January 2012 – Traffic; 17 July 2012 – Nucleation.)

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15, 10123–10162, 2015

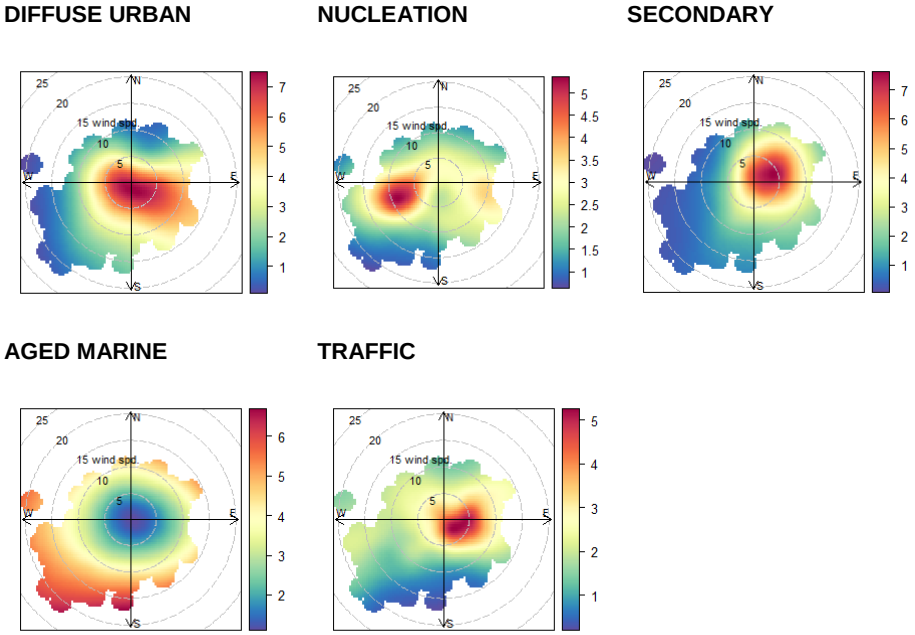

Figure 10. Polar plots showing how the PMF factors derived from the combined chemical composition/NSD dataset are affected by the daily vector average wind velocity and direction. (Units: G values (arbitrary units) and wind speed ($m\,s^{-1}$).)

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